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Cooling Performance Improvement of the Heat Driven Type Metal Hydride Refrigerator

-Heat Transfer Enhancement Influence of Metal Hydride Sheet Loading into a Metal Hydride Particle Bed-

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ABSTRACT

In the refrigeration and air conditioning fields, the demands of energy conservation and renewable energy have been increased recently. In this study, we aim at the development of the heat driven type metal hydride (abbr., MH) that can be driven by the low temperature exhaust or solar heat under 100°C. In order to use this system commercially, heat transfer enhancement of MH particle bed, activation characteristic improvement and production cost reduction of MH must be achieved. In this study, we use the two heat transfer enhancement methods for improving the low effective thermal conductivity of MH particle bed. One is the MH sheet and another is the brush type carbon fiber. MH sheet is inserted into MH layer. And, by this method, we aim not only to enhance the heat transfer of MH particle bed but also to achieve the temperature uniformity of MH particle one. The cooling performance of our MH refrigeration system is estimated by measurement and calculation.

1. INTRODUCTION

In order to reduce the CO₂ emission, we aim to develop the heat driven type MH water cooler. This water cooler is one of the chemical heat pumps, and the endothermic reaction of MH₂ is used for the cooling. The advantages of this system are shown in as follows: 1. Environmental pollution is quite low, for the working fluid of this system is hydrogen. 2. This system is noiseless and low vibration. 3. If we chose the proper MH alloys, we can use the low temperature exhausted heat and the solar heat as the heat source. But, this system is still not used commercially, because MH alloy is too expensive (200~300 \$ per 1kg MH) and has a low heat transfer performance and a bad activation characteristic.

For this problem, referring past research, Nagel et al. (1986) introduced fin inside MH particle bed, Suda et al. (1991) inserted copper mesh in MH particle bed, Takeda et al (1997) inserted metals particles with high thermal conductivity into MH particle bed, etc. are proposed, but system is getting heavy and complicated. For our research, we inserted MH sheet composed of MH alloy, carbon fiber as heat promoter, and fine fiber for sheet reinforcement to enhance heat transfer of MH particle bed. This MH sheet is new heat promoter. By enclosing MH sheet inside heat exchanger of MH refrigerator, it is assumed to improve heat transfer without reducing enclosed rate of MH.

2. EXPERIMENTAL APPARATUS

2.1 MH Alloys

The MH alloy compositions used in this study are shown in Table 1.

Table 1: MH Alloy Composition

MH1	$\text{TiFe}_{0.9}\text{Ni}_{0.1}$
MH2	$\text{La}_{0.6}\text{Y}_{0.4}\text{Ni}_{4.9}\text{Al}_{0.1}$

Also, these alloys are produced by Hokkaido University for this research. The alloys are produced by combustion synthesis method, enabling the activation readily. The metal used for the alloy is chosen by its cost, but still have high hydrogen absorption volume per unit mass.

2.2 MH Sheet

In this study, we compared the effective thermal conductivity of the MH layer with MH sheet enclosed inside. We changed the volume ratio of MH sheet, and the composition of MH sheet. The MH alloy contained inside the MH sheet has same composition corresponding to the alloy enclosed together. The photograph and the micrograph of MH sheet are shown in Figure 1. The sheet shown in Fig.1 is for MH1, and sheet for MH2 have same appearance.

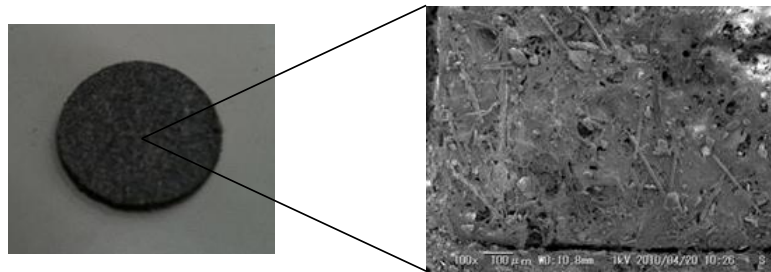


Figure 1: Photograph and micrograph of MH Sheet

2.3 Specification of Heat Exchanger

This heat exchanger is made of SUS316 circular pipe with external diameter of 19.05 mm and internal diameter of 15.75 mm. To endure the internal pressure of 3 MPa, the thickness of the pipe is 1.65 mm. Sintered metallic filter is connected above the heat exchanger. This filter is installed to prevent outflow of pulverized MH particle inside the heat exchanger during vacuum and entry of foreign matter to heat exchanger during hydrogen loading. Filter vent size is 0.5 μm . To measure the temperature history during refrigeration and regeneration process at the center of the MH particle bed inside the heat exchanger, 1mm diameter T type mineral insulated thermocouple (accuracy of $\pm 0.1^\circ\text{C}$) is inserted in the center of heat exchanger at height of 70 mm as shown in Figure 2. At the same height, T type thermocouple wire (accuracy of $\pm 0.1^\circ\text{C}$) is placed at the surface of heat exchanger to measure the temperature history of heat exchanger surface. This heat exchanger is used at both the effective thermal conductivity measurement and small scale model system. But, for the effective thermal conductivity measurement, heat exchanger is made of copper. The copper made heat exchanger have mostly same measurement as the heat exchanger made of SUS316 shown in Figure 3. Only that the ambient gas will enter from the thermocouple side.

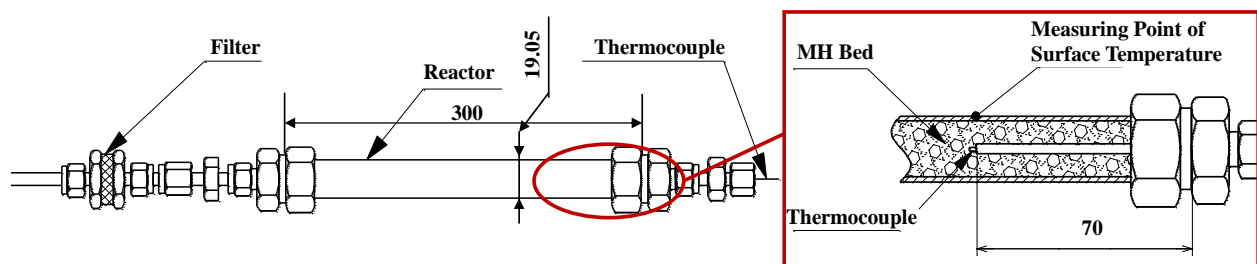


Figure 2: Specification of Heat Exchanger

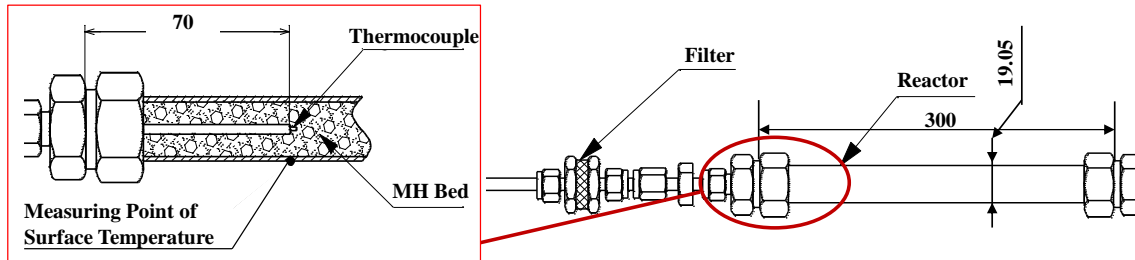


Figure 3: Specification of Heat Exchanger (Copper)

2.4 Small Scale Model System

Fig. 4 shows the model system apparatus. It consists of the heat exchangers of MH1 and MH2, the measurement system, the hydrogen supply and exhaust part, and three constant temperature baths for heat source, heat sink, and cooling load. In order to estimate the cooling load, we measure the accumulated hydrogen mass of the model system in various conditions. The thermal type mass flow meter (accuracy of $\pm 1\%$, maximum flow of 15NL/min), which is installed in the place between the MH1 and MH2 heat exchanger is used to measure the accumulated hydrogen mass. And, the T-type mineral insulated thermo-couples which are set up to the centers of MH1 and MH2 beds and constant temperature baths are used to measure the temperature change history. In addition, we install the pressure gauge (accuracy of $\pm 0.005\sim 0.008\%$) of full scale between MH1 heat exchanger and MH2 one, and measure the pressure change history. Silicon oil is used as working fluid of MH1, and water is used as working fluid of MH2.

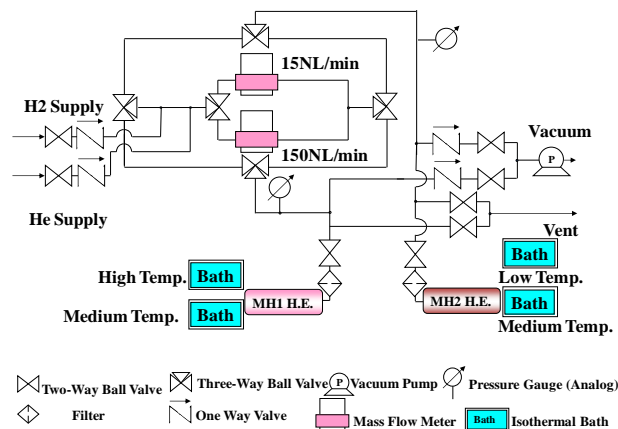


Figure 4: Small Scale Model System

3. EXPERIMENTAL METHOD

Our experimental procedure of model system is taken as follows:

① Activation and Stabilization Procedure

At first, MH alloy particles are charged into the heat exchanger, and are activated and stabilized. 2 mass% of MH sheet is enclosed inside the MH alloy particles bed to enhance the heat transfer. MH sheet is enclosed equally inside the heat exchanger.

② Hydrogen Supplying

We supply the hydrogen to heat exchangers of MH1 and MH2. And then, MH1 heat exchanger is immersed into the constant-thermal bath which is set at the heat source temperature, 125 °C and MH2 heat exchanger is immersed into the constant-thermal bath which is set at the heat sink temperature, 15 °C. Next, this system is vacuumed. After evacuating, we supply again the hydrogen of which pressure is controlled to 1MPa to the system. During hydrogen supplying, MH1 and MH2 alloys absorb the hydrogen. The temperatures of MH alloy particle bed are increased by this hydrogen absorption (exothermic reaction). We wait for the exothermic reaction to end. Reaction finish has been judged by measuring the temperature change of MH alloy bed. When the MH alloy temperatures reach the

setting temperature of heat media, we stop supplying the hydrogen.

③ Refrigeration Process Measurement.

After taking up the measurement, the MH1 heat exchanger is held up, and is immersed into the constant thermal bath which temperature is set to the heat sink temperature, 15 °C. This temperature change of MH1 heat exchanger is the trigger of MH1 hydrogen absorbing. And, by this MH1 hydrogen absorbing, MH2 starts to desorb the hydrogen. When the hydrogen flow rate of this system stop changing (under 0.01NI/min) the measurement of the experiment will be finished.

④ Regeneration Process Experiment.

We replace the MH1 heat exchanger on the constant thermal bath which is set at heat source temperature, 125 °C and, take up the measurement again. This temperature change of MH1 heat exchanger is the trigger of MH1 hydrogen desorbing. And, MH2 begins to absorb the hydrogen. When the hydrogen flow rate of this system stop changing (under 0.01NI/min) the measurement of the experiment will be finished.

⑤ One cycle of this system

Heat driven type MH refrigerator is composed of the two sets of heat exchangers (two MH1 and two MH2). In order to obtain the continuous cooling load, one set is on regeneration process while another one is on refrigeration process. Then, one cycle of this system consists of one refrigeration process and one regeneration process. We operate and measure the three cycles in this study to confirm that the activation of the MH alloy was done properly. And, the longer process between the refrigeration process and the regeneration process determine the half cycle time and the cycle time of this system.

4. CALCULATION METHOD

In order to estimate the influence of MH sheet given to the performance of MH heat exchanger, the unsteady one-dimensional cylindrical coordinate MH alloy particle bed model is built and calculated. Assumptions of our calculation are as follows.

- Heat transfer in the MH particle bed is done by only heat conduction.
- External wall of heat exchanger is insulated.
- Thermal properties do not depend on the temperature.
- MH alloy is filled in the heat exchanger without space.
- There is no pressure loss inside the heat exchanger.
- The physical properties of MH alloy are after activation.

The model of this calculation, shown in Figure 5, is assumed to be the cylindrical immersing type heat exchanger of the small scale model system.

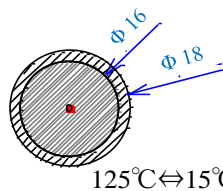


Figure 5: Calculation Model

In this calculation, the reaction heat of MH is obtained from the Van't Hoff equation. The exothermic reaction heat (MH1) is 33.84 k J/molH₂ and the endothermic reaction heat (MH1) is -37.40 k J/molH₂.

The thermal conductivity equation of unsteady one-dimensional cylindrical coordinate that uses to our calculation is shown in equation 1.

$$\frac{\partial T}{\partial t} = a \left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} \right) + \frac{Q}{c\rho} \quad \left(a = \frac{\lambda}{c\rho} \right) \quad (1)$$

Reaction heat per volume is calculated using equation 2.

$$Q = \frac{Y \cdot \rho_d}{2.016} \times H \times \frac{1}{\Delta T} \quad (2)$$

And, equation 1 is changed to temperature finite difference equation 3 using the full-implicit method.

$$T_i^{k+1} = \frac{1}{1+2c_r} \left[c_r \left\{ \left(1 + \frac{\Delta r}{2r_i} \right) T_{i+1}^{k+1} + \left(1 - \frac{\Delta r}{2r_i} \right) T_{i-1}^{k+1} \right\} + T_i^k + 2c_q Q_i \right] \quad \left(c_r = \frac{a\Delta t}{\Delta r^2} \quad c_q = \frac{\Delta t}{c\rho} \right) \quad (3)$$

5. EXPERIMENTAL AND CALCULATION RESULTS AND CONSIDERATION

5.1 Effect of MH Sheet on Effective Thermal Conductivity

To see the effect of MH sheet on effective thermal conductivity, we conducted various measurements. The basic experimental conditions used through all the measurements of effective thermal conductivity are shown in Table 2. The final data of these experiments are the average of 3 taken data.

Table 2: Common Experimental Condition

Mass of Alloy g	100
Ambient Gas	He
Diameter of MH Sheet mm	15
Pressure MPa	0, 0.1, 0.2, 0.4, 0.6, 0.8, 1.0

5.1.1 The effect of MH sheet on effective thermal conductivity: To see the effect of enclosing the MH sheet inside the heat exchanger, we first measured the effective thermal conductivity before and after enclosing the MH sheet inside the heat exchanger. MH sheets are enclosed inside the heat exchanger equivalently. The experimental conditions are shown in Table 3.

Table 3: Experimental Condition (Effect Verification)

			MH1	MH2
Number of MH Sheet			0, 20	
Volume Ratio of MH Sheet vol. %			0, 5.2	
Void Ratio %	Metal Hydride		7.0	6.8
	Carbon Fiber		5.7	5.9
	Fine Fiber	Material	Aramid JSP	
		Volume Ratio	4.6	4.7
Bulk Density kg/m ³			500.0	484.8
Thickness mm			0.78	
MH Sheet No. Assignment			MH1-2	MH2-2

The results of this experiment are shown in Figure 5.

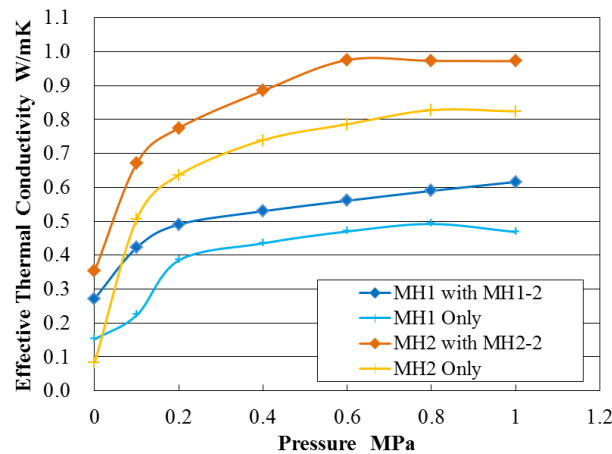


Figure 6: Effect of MH Sheet on Effective Thermal Conductivity

From Figure 6, the improvement of effective thermal conductivity can be seen. The effective thermal conductivity of MH1 improved 1.32 times and MH2 improved 1.18 times by enclosing MH sheet. From this result, MH sheet is effective to improve the heat transfer of the heat exchanger. Also, by enclosing the MH sheet inside heat exchanger, MH sheet act as cushion and prevent MH particle bed from consolidating.

5.1.2 Effective thermal conductivity comparison by the number of MH Sheet: We conducted effective thermal conductivity comparison by the number of MH to see the optimal amount to enclose inside the heat exchanger. The experimental conditions are shown in Table 4.

Table 4: Experimental Conditions (Number Change)

		MH1
Number of MH1-2 Sheet		0, 10, 20, 30, 40, 50
Volume Ratio of MH Sheet vol. %		0, 2.6, 5.2, 7.8, 10.4, 13.0
Composition vol. %	Metal Hydride	7.0
	Carbon Fiber	5.7
	Aramid Fiber	4.6
Bulk Density kg/m^3		500.0
Thickness mm		0.78

The results of this experiment are shown in Figure 7.

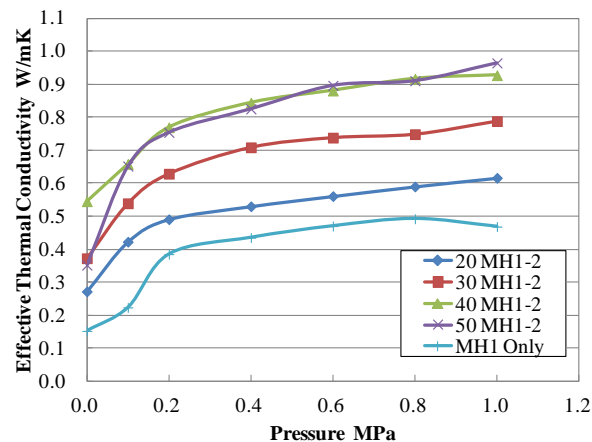


Figure 7: Effective Thermal Conductivity Comparison by the Number of MH1-2 Sheet (MH1)

As you can see from Figure 7, for this experimental condition, the number of MH sheet optimal for enclosing inside the heat exchanger is 40, with effective thermal conductivity of 0.93 W/mK at 1 MPa. When we enclosed 50 MH sheet, the effective thermal conductivity became similar with the effective thermal conductivity of 40 MH sheet. This show that the effective thermal conductivity will not improve much when over 40 MH sheet is enclosed inside the heat exchanger. From the result of this experiment, we concluded that the number of MH sheet optimal for the heat exchanger is 40. Comparing the effective thermal conductivity of before and after enclosing 40 MH sheet, 2.06 times of improvement was conformed.

5.1.3 Effective thermal conductivity comparison by the composition of MH sheet: MH sheet is composed of MH alloy, carbon fiber, and fine fiber. To see which parameter should be given priority, we measured the effective thermal conductivity by changing the composition of MH sheet. The experimental conditions are shown in Table 5.

Table 5: Experimental Condition (Composition Change)

		MH1			MH2		
Number of MH Sheet		20					
Volume Ratio of MH Sheet vol.%		5.2					
Sheet No. Assignment		MH1-1	MH1-2	MH1-3	MH2-1	MH2-2	MH2-3
Composition vol.%	Metal Hydride	7.9	7.0	5.6	7.9	6.8	5.6
	Carbon Fiber	2.9	5.7	9.1	2.9	5.9	9.1
	Aramid	4.7	4.6	4.7	4.7	4.7	4.7
Bulk Density kg/m ²		491.5	500.0	497.9	484.8	484.8	491.8
Thickness mm		0.70	0.78	0.78	0.69	0.82	0.79

The result of this experiment is shown in Figure 8.

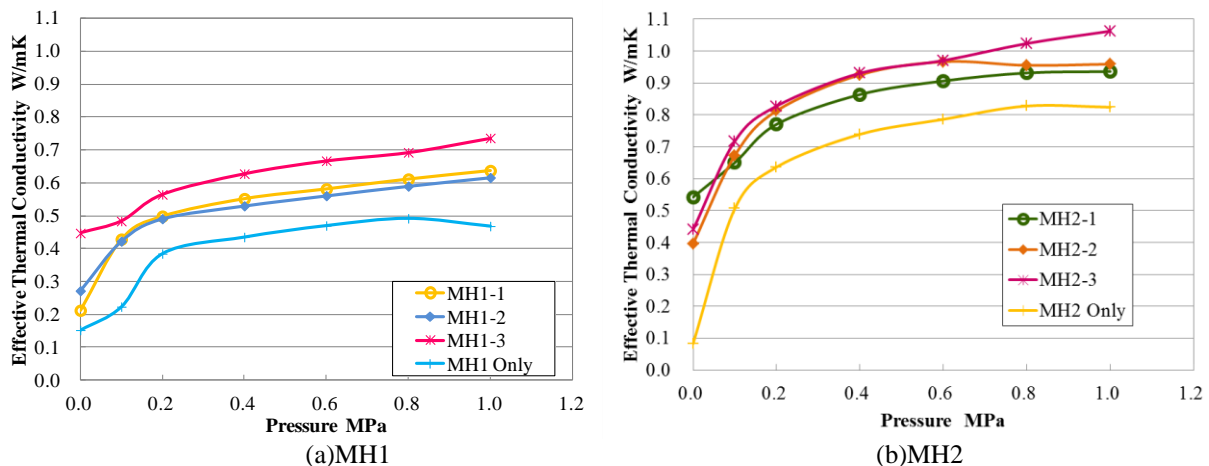


Figure 8: Effective Thermal Conductivity Comparison by the Composition of MH Sheet (MH1)

From Figure 8, you can see that the effective thermal conductivity is the highest when maximum carbon fiber is composed in MH sheet with 0.74 W/mK for MH1 and 1.06 W/mK for MH2. Since MH1-3 and MH2-3 are MH sheets with maximum carbon fiber composed inside, carbon fiber inside the MH sheet is assumed to give large influence to the effective thermal conductivity.

5.2 Small Scale Model System

From the experiment we have done so far, enclosing the MH Sheet inside the heat exchanger improves the effective thermal conductivity, showing that it is effective. For the next step, we enclosed MH sheet inside the heat exchanger of actual small scale model system to see the improvement of refrigeration capacity improvement. The experimental condition for this experiment is shown in Table 6. The MH sheet for this experiment is same as previous experiment. So the MH sheet specification is shown in Table 5. Also we named each run as Run 1 (MH1-1 and MH2-1), Run 2

(MH1-2 and MH2-2), and Run 3 (MH1-3 and MH2-3).

Table 6: Experimental Condition (Small Scale Model System)

	MH1	MH2
Mass of Alloy g	150	100
	20	
MH Sheet vol.%	5.2	
Heat Source Temperature °C	125	
Heat Sink Temperature °C	15	
Cooling Temperature °C	15	
Initial H ₂ Pressure MPa	1.0	
Heat Source	Silicon Oil	Water
Half Cycle Time	All Hydrogen Moved (under 0.01NI/min)	

The temperature history comparison is shown in Figure 9, the accumulated H₂ volumes are shown in Figure 10, and the refrigeration capacity comparison is shown in Figure 11.

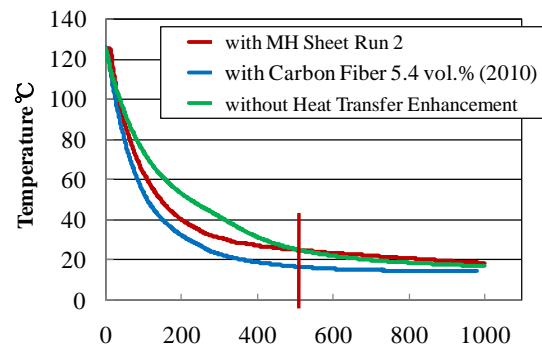


Figure 9: Temperature History Comparison between MH Sheet and Carbon Fiber (Refrigeration Process)\

We enclosed brush type carbon fiber (5.4 vol.%) as heat promoter last year enabling us to compare the data between carbon fiber and MH sheet. From Figure 9, from the heat promoting point of view, carbon fiber is more effective. But, the weight of MH sheet is 88% of carbon fiber, and the cost is 10% of carbon fiber. From this result, we concluded that MH sheet can be the alternative of carbon fiber.

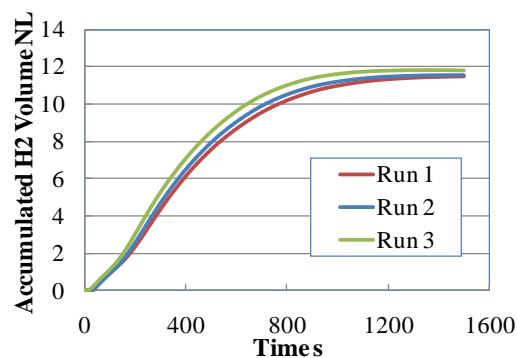


Figure 10: Accumulated H₂ Volume Comparison (Regeneration Process)

The effect of changing the composition of MH sheet was also confirmed from the accumulated H₂ volume on Figure 10. You can see that Run 3 finished the reaction faster than Run 1 and Run 2, resulting that MH sheet with maximum carbon fiber improved the heat transfer the most.

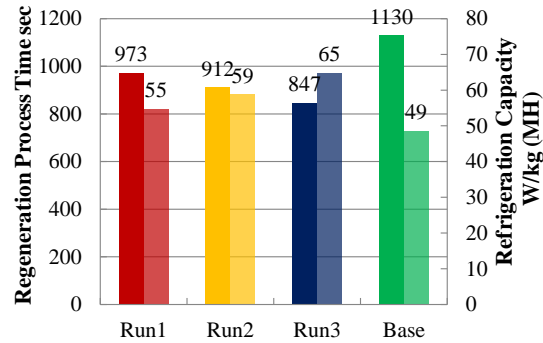


Figure 11: Refrigeration Capacity Comparison

The refrigeration capacity of MH refrigerator depends on half cycle time which is decided by longer process. For this experiment, it is regeneration process time. About Figure 11, for each run, the left data is regeneration process time, and the right data is refrigeration capacity calculated from the regeneration process time and alloy reaction heat. From Figure 11, by enclosing the MH sheet 5.2 vol%, half cycle time was reduced 25% at maximum. The refrigeration capacity calculated from half cycle time reduced 1.33 times at maximum. This result shows that the MH sheet with maximum carbon fiber inside is the most effective MH sheet for refrigeration capacity improvement.

5.3 Calculation Results

The comparison between the experimental result and calculation are shown in Figure 12.

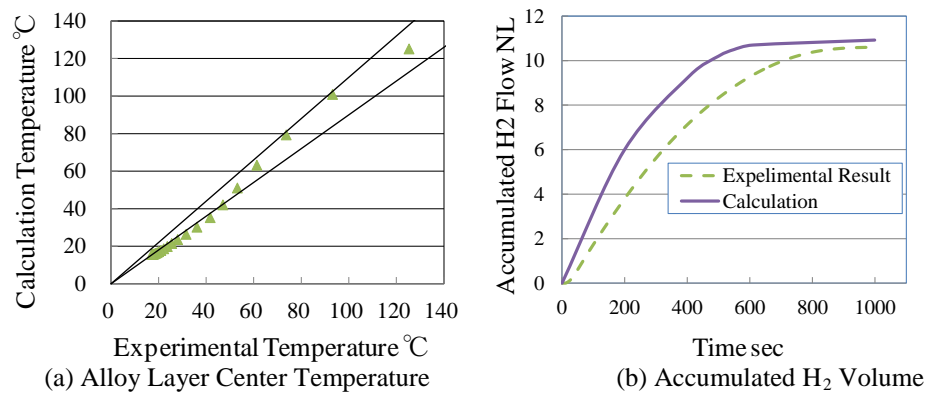


Figure 12: Comparison between Experimental Result and Calculation

From Figure 12 (a), the maximum error was 17%, and the average error was 10%, enabling us to describe the temperature history quantitatively. Using this temperature history, we calculated accumulated H₂ volume shown in Figure 12 (b). There is 30% error at maximum and needs to be improved. This can be done by taking the mesh more in detail. The refrigeration capacity calculated from the experimental value was 123 kJ/kg(MH), and the refrigeration capacity calculated from the calculated value was 127 kJ/kg(MH).

6. CONCLUSION

- By enclosing the MH sheet inside the heat exchanger, effective thermal conductivity improved 1.32 times at maximum.
- By enclosing 40 MH sheet to the refrigeration system, refrigeration capacity improved 2.06 times at maximum compared to the refrigeration capacity without MH sheet.
- The influence of alloy inside MH sheet is small, resulting that MH sheet is effective as heat promoter more than improvement of alloy enclose rate.
- From calculation result, temperature history average error was about 10% and refrigeration capacity became 127 kJ/kg(MH).

NOMENCLATURE

a	temperature conductivity	(mm ² /s)
c	specific heat	(-)
H	exothermic or exothermic enthalpy of MH	(-)
T	temperature	(°C)
Y	experimental transferred hydrogen amount	(NL)
λ	thermal conductivity	(-)
ρ	density	(kg/m ³)
ρ_d	bulk density	(kg/m ³)

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